

Spin wave approach to the two-magnon Raman scattering in an $J_{1x} - J_{1y} - J_2 - J_c$ antiferromagnetic Heisenberg model

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We study the two-magnon non-resonant Raman scattering in the (π, π) and $(\pi, 0)$ ordered antiferromagnetic phases of a $J_{1x} - J_{1y} - J_2 - J_c$ Heisenberg model on the tetragonal lattice within the framework of the spin-wave theory. We discuss the effects of various tuning factors to the two-magnon Raman spectra. We find that both the magnetic frustration J_2/J_1 and the interlayer exchange coupling J_c may significantly affect the spectra in both the B_{1g} and A'_{1g} channels in the (π, π) Néel ordered phase. Moreover, we find a splitting of the two-magnon peak in the $(\pi, 0)$ antiferromagnetic phase. We further discuss the implications of our results to the BaMnBi₂ and iron pnictide systems.

I. INTRODUCTION

In the recent years, the discovery of iron-based superconductors have triggered tremendous research in this new class of high T_c superconductors. Similar to cuprates, iron-based superconductors have a layered structure, and their parent compounds have long-range antiferromagnetic order. Superconductivity emerges when the magnetic order is destroyed by doping. It is now believed that magnetism is crucial for superconductivity in these materials. Typically the parent compounds of iron pnictides have a $(\pi, 0)$ collinear antiferromagnetic order. The magnetic properties of these materials can be well captured by the strong coupling approaches involving interactions of Fe local spins, described by effective $J_1 - J_2$ like models. These extended antiferromagnetic Heisenberg models are widely used to explain the magnetic properties of parent iron pnictides¹.

There are many other materials which invoke the local moment models. Recently, a class of novel manganese based materials AMnBi₂ ($A = \text{Sr, Ca}$) have attracted considerable research interests for their coexistence of itinerant Dirac electrons and long-range magnetic order associated with local moments. These materials share similar structural and electronic properties to iron pnictides. Insulating MnBi layer with Néel-type antiferromagnetic order on each Mn site² and ABi layer accommodating highly anisotropic Dirac carriers²⁻¹⁰ are alternatively stacked. These materials have provided an opportunity to explore the interplay of magnetism and Dirac itinerant carriers.

In studying the magnetic properties of these systems, Raman scattering is a powerful spectroscopic technique. It probes two-magnon correlations in which short-wavelength excitations dominates. The standard magnetic Raman scattering theory is based on the Fleury-Loudon coupling between the light and the spin system¹¹. Such a theory can be derived in the large- U Hubbard model at half-filling in the non-resonant regime¹². Near resonance where the incoming photon frequency is close to the band gap value, FL theory fails as the charge transfer process becomes dominant¹³.

Even in the non-resonant regime, theoretical understanding of Raman scattering in spin systems is still very limited. Early works on the simple 2D antiferromagnets have revealed that magnon-magnon (m-m) interactions have significant influence on the shape of Raman

spectra as multiple scattering of magnon pairs excited by photons is non-negligible in the Raman process¹⁴. Magnetic Raman scattering in 2D simple antiferromagnets has been further studied using various approaches: spin wave & Green's function theory¹⁵⁻¹⁷, Exact Diagonalization (ED), Quantum Monte Carlo method¹⁶, etc.. Ref.¹⁵ claimed that four-magnon intensity is too small compared with two-magnon ones.¹⁷ calculated both two-magnon and four magnon Raman spectra for a 2D frustrated $J_1 - J_2$ systems with Néel order using the modified spin wave (MSW) theory. However, their calculations were still at the mean-field level, which ignored higher order scattering processes of magnon pairs. Ref.¹⁸ calculated 2D systems with ring exchange interactions. Ref.¹⁹ launched ED calculations for specific iron-based materials in $(\pi, 0)$ collinear and $(\pi/2, \pi/2)$ diagonal double stripe order. However, their calculations were restricted to small spin ($S \leq 1$), small cluster sizes ($N_c \leq 36$) and very limited system parameters.

Up to now, a convincing and detailed work that is applicable to systems with frustration, exchange anisotropy, and finite interlayer exchange couplings is still absent. In this article we present a systematic study of the two-magnon non-resonant Raman scattering in (π, π) and $(\pi, 0)$ ordered antiferromagnets with square/tetragonal lattice geometry (notice that the magnetic symmetry can be lower than lattice symmetry) within the framework of spin-wave theory. We will discuss its general feature, and implications of spin magnitude, frustration, anisotropy and interlayer coupling to Raman spectra. The article is organized as follows. In Sec. II we introduce our calculation method. In Sec. III and IV we present our results in (π, π) and $(\pi, 0)$ ordered system respectively. Finally in Sec. IV we present our discussions and concluding remarks.

II. GENERAL FORMALISM

We study a spin- S $J_{1x} - J_{1y} - J_2 - J_c$ Heisenberg model on a tetragonal lattice. The Hamiltonian reads

$$H = \frac{J_{1x}}{2} \sum_{i, \delta_x} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta_x} + \frac{J_{1y}}{2} \sum_{i, \delta_y} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta_y} + \frac{J_2}{2} \sum_{i, \delta_x, \delta_y} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta_x+\delta_y} + \frac{J_c}{2} \sum_{i, \delta_z} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta_z} \quad (1)$$

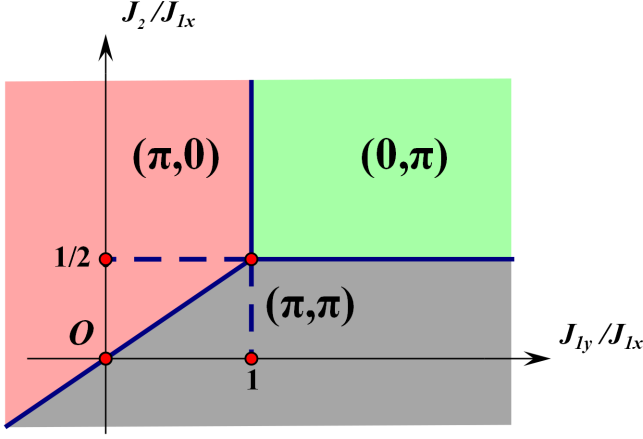


Figure 1. (Color online). Ground state phase diagram of the classical $J_{1x} - J_{1y} - J_2$ model in the regime $J_{1x} > 0$, $J_{1y}/J_{1x} \geq -1$ and $J_{1y} + 2J_2 \geq 0$, where the ground state stabilizes in-plane AFM orders with wave vectors $(\pi, 0)$, $(0, \pi)$, and (π, π) , respectively. The solid lines mark the phase boundaries.

where \mathbf{S}_i refers to a spin at lattice site i , and $\delta_x = \pm a\hat{x}$, $\delta_y = \pm a\hat{y}$, $\delta_z = \pm c\hat{z}$ are nearest neighbor vectors along x, y, z directions, respectively. J_{1x} , J_{1y} , J_2 , and J_c are, respectively, the exchange couplings between row and column nearest neighbors, next nearest neighbors, and nearest interlayer neighbors. In this paper, we are interested in the Raman scattering with an AF ground state. Without losing generality, we take $J_{1x} > 0$ in this model.

The ground state phase diagram of the classical $J_{1x} - J_{1y} - J_2$ model is illustrated in Fig. 1. In the regime shown, three in-plane AFM ground states can be stabilized. In each of the three ordered states, the quantum fluctuations of the corresponding quantum spin model are taken into account by the spin-wave approach via a standard $1/S$ expansion, which is expected to be a good approximation when the system is not in the vicinity of the classical phase boundaries.

A. $1/S$ Expansion

By introducing Holstein-Primakoff (H-P) transformation on the bipartite tetragonal lattice, we express spins in the A (spin up) and B (spin down) sublattices in terms of bosonic operators a_l and b_m

$$\begin{aligned} S_l^z &= S - a_l^\dagger a_l \\ S_l^+ &= \sqrt{2S} f_l(S) a_l \\ S_m^z &= -S + b_m^\dagger b_m \\ S_m^+ &= \sqrt{2S} b_m^\dagger f_m(S), \end{aligned} \quad (2)$$

where

$$f_{l/m}(S) = \sqrt{1 - \frac{n_{l/m}}{2S}}, \quad (3)$$

$n_{l/m} = a_{l/m}^\dagger a_{l/m}$, and $l \in A$ and $m \in B$. For Eq. (3) to be valid, the bosons must be restricted in the $n_{l/m} \leq 2S$ physical space.

Then we perform an $1/S$ expansion for $f_{l/m}(S)$ up to the $1/S$ order:

$$f_{l/m}(S) = 1 - \frac{n_{l/m}}{4S} + \dots, \quad (4)$$

and perform a Fourier transformation for the bosonic operators

$$a_l = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} a_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}_l}, \quad b_m = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} b_{-\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_m} \quad (5)$$

where \mathbf{k} is defined in the first Brillouin zone (FBZ) of the momentum space.

The Hamiltonian is also expanded in powers of $1/S$ as

$$H = E_0 + H_0 + H_1 + O(1/S^2). \quad (6)$$

Here, E_0 corresponds to the classical energy of the system. H_0 corresponds to the quadratic linear spin wave (LSW) terms, which takes the form

$$H_0 = \sum_{\mathbf{k}} P_{\mathbf{k}} (a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + b_{-\mathbf{k}}^\dagger b_{-\mathbf{k}}) + Q_{\mathbf{k}} (a_{\mathbf{k}}^\dagger b_{-\mathbf{k}}^\dagger + a_{\mathbf{k}} b_{-\mathbf{k}}), \quad (7)$$

where the coefficients $P_{\mathbf{k}}$ and $Q_{\mathbf{k}}$ are defined in the Appendix. Next we perform Bogoliubov transformation

$$a_{\mathbf{k}}^\dagger = l_{\mathbf{k}} \alpha_{\mathbf{k}}^\dagger + m_{\mathbf{k}} \beta_{-\mathbf{k}}, \quad b_{-\mathbf{k}} = m_{\mathbf{k}} \alpha_{\mathbf{k}}^\dagger + l_{\mathbf{k}} \beta_{-\mathbf{k}} \quad (8)$$

where $l_{\mathbf{k}} = \sqrt{\frac{1+\epsilon_{\mathbf{k}}}{2\epsilon_{\mathbf{k}}}}$, $m_{\mathbf{k}} = -x_{\mathbf{k}} l_{\mathbf{k}} = -\text{sgn} \gamma_{\mathbf{k}} \sqrt{\frac{1-\epsilon_{\mathbf{k}}}{2\epsilon_{\mathbf{k}}}}$, $\epsilon_{\mathbf{k}} = \sqrt{1 - \gamma_{\mathbf{k}}^2}$, $\gamma_{\mathbf{k}} = Q_{\mathbf{k}}/P_{\mathbf{k}}$. H_0 is then diagonalized as

$$H_0 = \sum_{\mathbf{k}} \omega_{\mathbf{k}} (\alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}} + \beta_{-\mathbf{k}}^\dagger \beta_{-\mathbf{k}} + 1) - P_{\mathbf{k}}, \quad (9)$$

where $\omega_{\mathbf{k}} = P_{\mathbf{k}} \epsilon_{\mathbf{k}}$.

H_1 corresponds to the $1/S$ order correction to the LSW results. It is written in Bogoliubov magnons as

$$H_1 = \text{const.} + H'_0 + H'_1 + \dots \quad (10)$$

where

$$H'_0 = \sum_{\mathbf{k}} A_{\mathbf{k}} (\alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}} + \beta_{\mathbf{k}}^\dagger \beta_{\mathbf{k}}) + B_{\mathbf{k}} (\alpha_{\mathbf{k}}^\dagger \beta_{-\mathbf{k}}^\dagger + \alpha_{\mathbf{k}} \beta_{-\mathbf{k}}), \quad (11)$$

is known as the Oguchi correction arising from transforming the bosonic operators into normal products. The Oguchi terms give the $1/S$ order correction to the magnon dispersion $\tilde{\omega}_{\mathbf{k}} = \omega_{\mathbf{k}} + A_{\mathbf{k}}$.

$$\begin{aligned} H'_1 = \frac{2}{N} \sum_{1234} \delta_{\mathbf{G}} (1 + 2 - 3 - 4) l_1 l_2 l_3 l_4 [& B_{1234}^{(1)} \alpha_1^\dagger \alpha_2^\dagger \alpha_3^\dagger \alpha_4^\dagger + \\ & B_{1234}^{(2)} \beta_{-3}^\dagger \beta_{-4}^\dagger \beta_{-1} \beta_{-2} + B_{1234}^{(3)} \alpha_1^\dagger \beta_{-4}^\dagger \beta_{-2} \alpha_3 + \\ & (B_{1234}^{(4)} \alpha_1^\dagger \beta_{-2} \alpha_3 \alpha_4 + B_{1234}^{(5)} \beta_{-4}^\dagger \beta_{-1} \beta_{-2} \alpha_3 + \\ & B_{1234}^{(6)} \alpha_1^\dagger \alpha_2^\dagger \beta_{-3}^\dagger \beta_{-4}^\dagger + h.c.)] \end{aligned} \quad (12)$$

where 1, 2, 3, 4 are abbreviations of the wave vectors \mathbf{k}_1 , \mathbf{k}_2 , \mathbf{k}_3 , \mathbf{k}_4 , which are also defined in FBZ, $\delta_{\mathbf{G}} (1 + 2 - 3 - 4)$ represents the conservation of momenta within a reciprocal lattice vector \mathbf{G} . H'_0 consists of two-magnon scattering terms of the magnon-magnon (m-m) interaction depending on the coefficients $P_{\mathbf{k}}$, $Q_{\mathbf{k}}$, $A_{\mathbf{k}}$, $B_{\mathbf{k}}$ and the vertex factor $B_{1234}^{(3)}$, whose explicit forms are given in Appendix A.

B. Two-magnon Raman operator

In the standard magnetic Raman FL theory, the second order Raman scattering operator is given by¹¹

$$\hat{O} = \frac{\lambda}{2} \sum_{ij} J_{ij} (\hat{\mathbf{e}}_{in} \cdot \hat{\mathbf{d}}_{ij}) (\hat{\mathbf{e}}_{out} \cdot \hat{\mathbf{d}}_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j \quad (13)$$

where $\hat{\mathbf{e}}_{in}$ and $\hat{\mathbf{e}}_{out}$ are unit polarization vectors of the incoming and scattered lights. $\hat{\mathbf{d}}_{ij}$ is the vector connecting site i and site j . λ is the coupling constant, which is scaled to $\sqrt{\frac{2}{N}}$, as its magnitude is not important for our results.

We will consider the following light polarization geometries which are widely accepted in experimental set up: $\hat{\mathbf{e}}_{in} = \frac{1}{\sqrt{2}}(\hat{x} + \hat{y})$, $\hat{\mathbf{e}}_{out} = \frac{1}{\sqrt{2}}(\hat{x} - \hat{y})$ for $x'y'$ polarization, and $\hat{\mathbf{e}}_{in} = \frac{1}{\sqrt{2}}(\hat{x} + \hat{y})$, $\hat{\mathbf{e}}_{out} = \frac{1}{\sqrt{2}}(\hat{x} + \hat{y})$ for $x'x'$ polarization. Here x' and y' refer to the rotated axes after a 45° rotation about the z axis in the xy plane.

Note that if the system has D_{4h} symmetry, $x'y'$ polarization corresponds to $B_{1g} \oplus A_{2g}$ symmetry group representations, and is usually called the B_{1g} channel because the A_{2g} component is zero in the second-order Raman scattering. The $x'x'$ polarization corresponds to $A_{1g} \oplus B_{2g}$ representations and is denoted as the A'_{1g} channel. In D_{4h} symmetric system these two channels are well separated from a symmetry perspective. If the D_{4h} symmetry is broken, they may not refer to the different irreducible representations of the symmetry group, and as a consequence, the scattering signals from these two channels can mix.

At the LSW level, the two-magnon part of the Raman operator is given by

$$\hat{O} = \sum_{\mathbf{k}} M_{\mathbf{k}} (\alpha_{\mathbf{k}}^\dagger \beta_{-\mathbf{k}}^\dagger + \alpha_{\mathbf{k}} \beta_{-\mathbf{k}}) + \dots \quad (14)$$

where explicit forms of $M_{\mathbf{k}}$ in B_{1g} and A'_{1g} channels are given in Appendix A.

C. Raman Scattering Cross Section

The Raman scattering cross section at zero temperature is given by $R(\omega) = -\frac{1}{\pi} \text{Im}[I(\omega)]$, where $I(\omega)$ is the correlation function $I(\omega) = -i \int dt e^{i\omega t} \langle \mathcal{T}_t \hat{O}^\dagger(t) \hat{O}(0) \rangle_0$. Here $\langle \dots \rangle_0$ represents quantum mechanical average over the ground state, and \mathcal{T}_t is the time ordering operator. The two-magnon contribution to $I(\omega)$ can be written as $I(\omega) = \sum_{\mathbf{k}, \mathbf{k}'} M_{\mathbf{k}} \Pi_{\mathbf{k}\mathbf{k}'}(\omega) M_{\mathbf{k}'}$. Here we define two-magnon Green's function

$$\Pi_{\mathbf{k}\mathbf{k}'}(\omega) = -i \int dt e^{i\omega t} \langle \mathcal{T} \alpha_{\mathbf{k}}(t) \beta_{-\mathbf{k}}(t) \alpha_{\mathbf{k}'}^\dagger(0) \beta_{-\mathbf{k}'}^\dagger(0) \rangle_0 \quad (15)$$

To calculate $I(\omega)$ we have adopted the following two simplifications^{15,20}:

1) We expand the one-magnon propagator up to the $1/S$ order. Given that there is no correction to the propagator at the $1/S$ order, they are identical to the unperturbed ones:

$$G_{\alpha\alpha}(\mathbf{k}, \omega) = G_{\beta\beta}(\mathbf{k}, \omega) = \frac{1}{\omega + i0^+ - \tilde{\omega}_{\mathbf{k}}} \quad (16)$$

$$G_{\alpha\beta}(\mathbf{k}, \omega) = G_{\beta\alpha}(\mathbf{k}, \omega) = 0$$

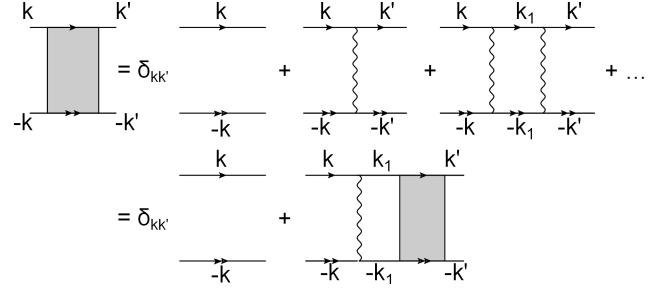


Figure 2. (Color online). Ladder diagrams for two-magnon propagator (15). A line with single arrow represents α magnon propagator $G_{\alpha\alpha}(\mathbf{k}, \omega)$. A line with double arrow represents β magnon propagator $G_{\beta\beta}(\mathbf{k}, \omega)$.

By applying Wick's theorem, the unperturbed two-magnon propagator can be expanded in terms of one-magnon ones as

$$\Pi_{\mathbf{k}\mathbf{k}'}^{(0)}(\omega) = \delta_{\mathbf{k}\mathbf{k}'} i \int \frac{d\omega_1}{2\pi} G_{\alpha\alpha}(\mathbf{k}, \omega + \omega_1) G_{\beta\beta}(-\mathbf{k}, -\omega_1)$$

$$= \delta_{\mathbf{k}\mathbf{k}'} \frac{1}{\omega + i0^+ - 2\tilde{\omega}_{\mathbf{k}}} \quad (17)$$

2) The m-m interaction is taken into account within the framework of the ladder approximation for the two-magnon Green's function (15). In this approximation, the core vertex is expanded up to the $1/S$ order, and only vertex terms equivalent to $\alpha^\dagger \beta^\dagger \beta \alpha$ are kept, as illustrated in Fig. 2. As the total momentum of incoming and outgoing α and β magnons are fixed to 0, the vertex is the function of the incoming and outgoing α magnon momentum \mathbf{k} and \mathbf{k}' , i.e. $\mathcal{V}_{\mathbf{k}\mathbf{k}'} = \frac{2}{N} B_{\mathbf{k}'\mathbf{k}\mathbf{k}\mathbf{k}'}^{(3)}$.

Therefore we have

$$\Pi_{\mathbf{k}\mathbf{k}'}(\omega) = i \int \frac{d\omega'}{2\pi} G_{\alpha\alpha}(\mathbf{k}, \omega + \omega') G_{\beta\beta}(-\mathbf{k}, -\omega') \Gamma_{\mathbf{k}\mathbf{k}'}(\omega, \omega') \quad (18)$$

where the vertex function $\Gamma_{\mathbf{k}\mathbf{k}'}(\omega, \omega')$ satisfies Bathe-Salpeter equation

$$\Gamma_{\mathbf{k}\mathbf{k}'}(\omega, \omega') = \delta_{\mathbf{k}\mathbf{k}'} + i \sum_{\mathbf{k}_1} \int \frac{d\omega_1}{2\pi} \mathcal{V}_{\mathbf{k}\mathbf{k}_1} G_{\alpha\alpha}(\mathbf{k}_1, \omega + \omega_1)$$

$$\times G_{\beta\beta}(-\mathbf{k}_1, -\omega_1) \Gamma_{\mathbf{k}_1\mathbf{k}'}(\omega, \omega_1) \quad (19)$$

Since $\mathcal{V}_{\mathbf{k}\mathbf{k}'}$ is independent of frequencies. From the equation (19), $\Gamma_{\mathbf{k}\mathbf{k}'}(\omega, \omega')$ is independent of ω' , i.e. $\Gamma_{\mathbf{k}\mathbf{k}'}(\omega, \omega') = \Gamma_{\mathbf{k}\mathbf{k}'}(\omega)$

In Eqs. (18) and (19), integration over frequencies are decoupled

$$\Pi_{\mathbf{k}\mathbf{k}'}(\omega) = \Pi_{\mathbf{k}\mathbf{k}}^{(0)}(\omega) \Gamma_{\mathbf{k}\mathbf{k}'}(\omega)$$

$$\Gamma_{\mathbf{k}\mathbf{k}'}(\omega) = \delta_{\mathbf{k}\mathbf{k}'} + \sum_{\mathbf{k}_1} \mathcal{V}_{\mathbf{k}\mathbf{k}_1} \Pi_{\mathbf{k}_1\mathbf{k}_1}^{(0)}(\omega) \Gamma_{\mathbf{k}_1\mathbf{k}'}(\omega) \quad (20)$$

Eliminating Γ vertex, we get two-magnon Dyson's equation:

$$\hat{\Pi} = \hat{\Pi}^{(0)} + \hat{\Pi}^{(0)} \hat{\mathcal{V}} \hat{\Pi} = \hat{\Pi}^{(0)} \sum_{n=0}^{+\infty} (\hat{\mathcal{V}} \hat{\Pi}^{(0)})^n \quad (21)$$

Directly solving such an equation rigorously $\hat{\Pi} = [\hat{1} - \hat{\Pi}^{(0)} \hat{\mathcal{V}}]^{-1} \hat{\Pi}^{(0)}$ would require the inverse of the matrix with a $N/2 \times N/2$ dimension, which is obviously computationally expensive. So we use the following alternative

approach: The vertex function can be expressed as a separable form

$$\mathcal{V}_{\mathbf{k}\mathbf{k}'} = \sum_{m,n=1}^{N_c} v_{m\mathbf{k}} \Gamma_{mn} v_{n\mathbf{k}} \quad (22)$$

where explicit forms of $\hat{\Gamma}$ and $v_{n\mathbf{k}}$ are given in Appendix A. We have

$$\begin{aligned} \hat{\Pi} &= \hat{\Pi}^{(0)} \sum_{n=0}^{+\infty} (\hat{v}^T \hat{\Gamma} \hat{v} \hat{\Pi}^{(0)})^n \\ &= \hat{\Pi}^{(0)} + (\hat{v} \hat{\Pi}^{(0)})^T \cdot \hat{\Gamma} \sum_{n=0}^{+\infty} [(\hat{v} \hat{\Pi}^{(0)} \hat{v}^T) \hat{\Gamma}]^n \cdot (\hat{v} \hat{\Pi}^{(0)}) \\ &= \hat{\Pi}^{(0)} + \hat{\Pi}^L \end{aligned} \quad (23)$$

where

$$\hat{\Pi}^L = (\hat{v} \hat{\Pi}^{(0)})^T \cdot \hat{\Gamma} [1 - (\hat{v} \hat{\Pi}^{(0)} \hat{v}^T) \hat{\Gamma}]^{-1} \cdot (\hat{v} \hat{\Pi}^{(0)}) \quad (24)$$

is the ladder correction to the two-magnon propagators.

Thus we have obtained an approach of exactly solving the Dyson's equation (21) with the price of inverting matrix with mere dimension of $N_c \times N_c$. Finally, the correlation function $I(\omega)$ can be obtained by

$$I(\omega) = I^{(0)}(\omega) + I^L(\omega) \quad (25)$$

where

$$I^{(0)}(\omega) = \hat{M}^T \hat{\Pi}^{(0)} \hat{M} \quad (26)$$

and

$$\begin{aligned} I^L(\omega) &= \hat{M}^T \hat{\Pi}^L \hat{M} = (\hat{v} \hat{\Pi}^{(0)} \hat{M})^T \\ &\cdot \hat{\Gamma} [1 - (\hat{v} \hat{\Pi}^{(0)} \hat{v}^T) \hat{\Gamma}]^{-1} \cdot (\hat{v} \hat{\Pi}^{(0)} \hat{M}) \end{aligned} \quad (27)$$

are non-interacting and ladder corrections to the total scattering cross section, respectively.

III. RESULTS FOR THE (π, π) NÉEL ORDER

We first consider the results of Raman scattering when the ground state has an AFM Néel order at wave vector (π, π) . We discuss several factors that may affect the Raman spectrum.

A. Role of $1/S$

We consider the effects of quantum fluctuations beyond the LSW level. To focus on this point, we limit our discussion to the case $J_{1x} = J_{1y} = J_1$ and $J_c = 0$ in this subsection. As is shown in Eq. (10), at the $1/S$ order, the corrections to the LSW results come from the following two parts: the Oguchi term H'_0 shifts the magnon dispersion to higher energies, and the m-m interaction term H'_1 allows repeat scattering of the light-excited magnon pairs. The roles of these two terms in two-magnon Raman spectra are shown in Fig. 3.

The non-interacting (LSW) spectrum of the B_{1g} channel typically shows a broad peak above an absorption edge at excitation energy $\omega \sim 4J_1$ (see Fig. 3(a) and (b)). When the m-m interaction is switched on, the spectral weight of this non-interacting part is suppressed, and an additional peak below the absorption edge is developed.

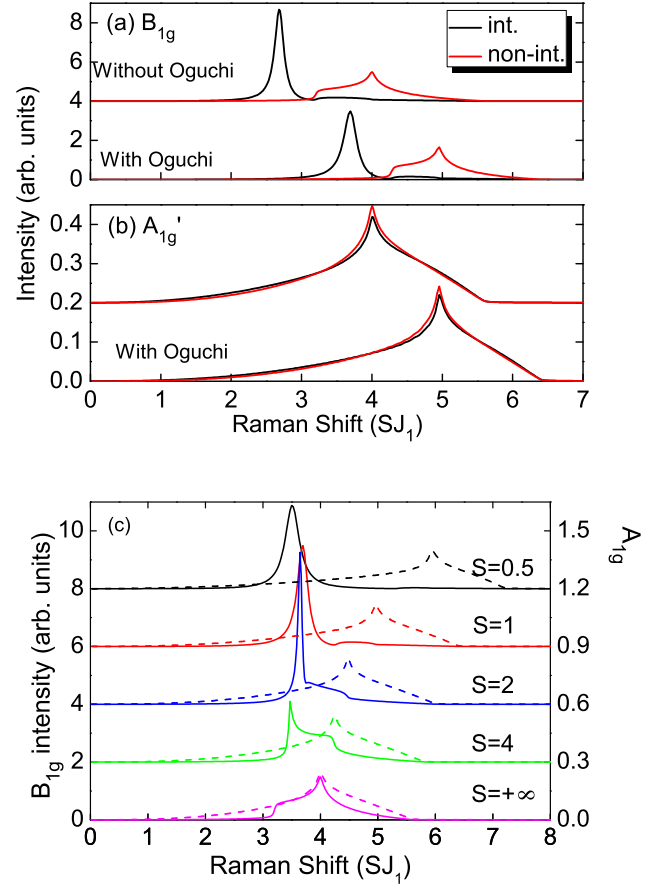


Figure 3. (Color online). Two-magnon Raman spectra for the $J_1 - J_2$ model with $SJ_1 = 1$, $SJ_2 = 0.3$. (a)(b): spectra for $S = 1$ highlighting the effects of Oguchi and m-m interaction terms. (c): dependence of S in B_{1g} (solid lines) and A'_{1g} (dashed lines) channels, both are calculated with the Oguchi and m-m interaction terms.

This peak is particularly sharp when the spin size $S \lesssim 4$, indicating a resonance feature in this channel (Fig. 3(c)). With increasing S , the position of the resonance peak is getting closer to the absorption edge, and its intensity is reduced, until eventually vanishes when $S \rightarrow \infty$. The existence of a sharp resonance peak makes the lineshape of the spectrum completely different once the m-m interaction is taken into account in the B_{1g} channel. On the other hand, for the A'_{1g} channel, the spectrum is almost not modified by the m-m interaction. In fact, as we will discuss below, the peak in this channel is associated with a van Hove singularity in the density of states (DoS) of the one magnon dispersion, not affected by the m-m interaction (see Fig. 4(c)(d)).

As shown in Fig. 3(a) and (b), the Oguchi term slightly changes the lineshape of the spectrum. Its main effect is to push the spectral weight to higher energy. This explains well the monotonic shift (toward higher energy) of the peak position in the A'_{1g} channel as decreasing S (Fig. 3(c)). While, for B_{1g} channel, the non-monotonic variance of the resonance peak position as decreasing S originates from the competition between the Oguchi term and m-m interactions.

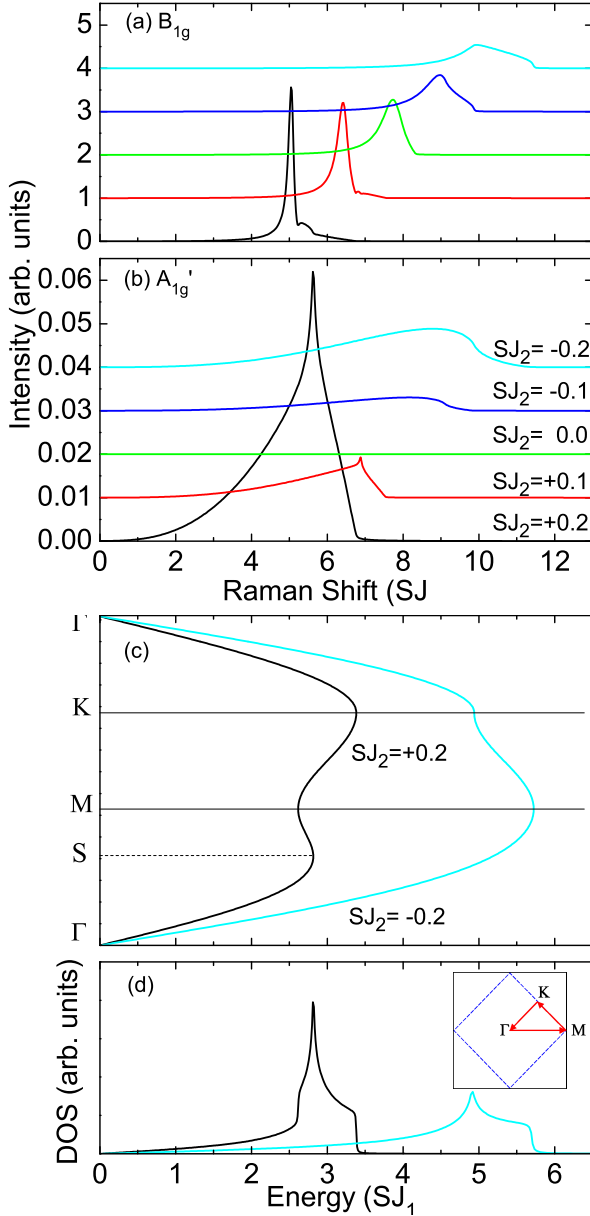


Figure 4. (Color online). (a)(b) Two-magnon Raman spectra for the frustrated $J_1 - J_2$ model $SJ_1 = 1$, $S = 2$ with different J_2 ; (c) magnon dispersion and (d) one magnon DoS for $SJ_1 = 1$, $SJ_2 = \pm 0.2$, $S = 2$.

B. Role of J_2 frustration

The next nearest neighbor exchange coupling J_2 introduces frustration to the ground state. To study the effects of frustration, we consider the frustrated $J_1 - J_2$ model with $SJ_1 = 1$ for various J_2 values. For $S = 2$, the two-magnon spectra in both B_{1g} and A'_{1g} channels are shown in Fig. 4. We see that in the frustrated models with increasing antiferromagnetic J_2 , the peaks of the Raman spectra in both channels become sharper, whereas in the non-frustrated models with ferromagnetic J_2 , the spectral weights only show very broad humps. This difference can be understood by examining the one-magnon dispersion curves for the frustrated and non-frustrated models, shown in Fig. 4(c), respectively. We find that the sharp resonance peak in the B_{1g} channel in the frustrated case comes from resonant scattering of magnon pairs $\alpha_{\mathbf{k}}$ and

$\beta_{-\mathbf{k}}$ near the $\mathbf{k} = (\pi, 0)$ (M) point, where the dispersion has a local minimum. The resonant peak is completely suppressed when the dispersion turns to a local maximum at the M point in the non-frustrated model.

The peak in the A'_{1g} channel, though evolves in a similar way as the resonance peak in the B_{1g} channel, has a very different origin. In the frustrated case, we find there exists four saddle points in the dispersion along the Γ -M line. One of them is labeled as the S point in Fig. 4(c). These saddle points contribute to a van Hove singularity of the one-magnon DoS with a logarithmic divergence. This van Hove singularity contributes to the sharp A'_{1g} peak. In the non-frustrated case, the saddle points along Γ -M line and the associated van Hove singularity are removed, and hence the sharp peak does not appear in the A'_{1g} channel. Note that besides the saddle points we just discussed, there can be two more (inequivalent) saddle points at $(\pi/2, \pi/2)$ and $(-\pi/2, \pi/2)$. But these saddle points do not contribute to singularities in the A'_{1g} channel.

C. Role of anisotropy

We now study the effect of the exchange anisotropy, $J_{1x} \neq J_{1y}$, on the Raman scattering. This anisotropy serves as another perturbation to the (π, π) AFM ground state. In the anisotropic $J_{1x} - J_{1y} - J_2$ model, the D_{4h} symmetry is reduced to D_{2h} . As discussed above, the B_{1g} and A'_{1g} channels will share components with same irreducible representations. In Fig. 5 we show the results for $SJ_{1x} = 1$, $SJ_{1y} = 0.9$, $SJ_2 = 0.3$, and $S = 2$. We see that the most significant changes of the spectrum by the anisotropy are in the A'_{1g} channel: First, an additional peak in the A'_{1g} channel emerges at the position of the resonance peak of the B_{1g} channel. This behavior clearly indicates the two channels are not well separated when the D_{4h} symmetry is broken. This peak is already visible when the anisotropy $J_{1y}/J_{1x} - 1 \gtrsim 5\%$. Therefore, it can be used to probe the possible exchange (and associated structural) anisotropy of materials as complementary to neutron diffraction. As another effect, the A'_{1g} peak in the isotropic model is split which reflects the anisotropy of the one-magnon dispersion along the $(0, 0)$ - $(\pi, 0)$ and $(0, 0)$ - $(0, \pi)$ directions.

D. Role of the interlayer exchange coupling

The previous results are discussed in 2D systems. In a more realistic 3D model, interlayer exchange coupling J_c will also affect the Raman spectra. Here we consider a $J_1 - J_2 - J_c$ model with $SJ_1 = 1$, $SJ_2 = 0.3$, and $S = 5/2$ for various J_c values. The results are shown in Fig. 6. We see that for both FM and AFM J_c , with increasing the magnitude of J_c , the sharp peak of the spectral weight in the A'_{1g} channel evolves to a very broad platform. One can prove that at the LSW level, the width of this platform is proportional to the magnitude of J_c .

In the B_{1g} channel it is remarkable that the sharp resonance peak feature is suppressed significantly by a FM J_c but preserves for an AFM J_c . Such a phenomenon can be understood as follows: when $J_c \neq 0$, the magnons are dispersive along the k_z direction. When $|J_c|$ is small, the magnon pair scattering term $B_{1234}^{(3)} \alpha_1^\dagger \beta_{-4}^\dagger \beta_{-2} \alpha_3$ has little

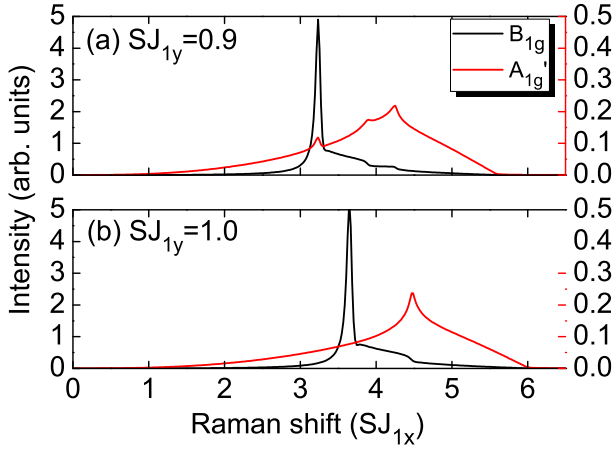


Figure 5. (Color online). Two-magnon Raman spectra for frustrated anisotropic $J_{1x} - J_{1y} - J_2$ model with $SJ_{1x} = 1$, $SJ_{1y} = 0.9$, $SJ_2 = 0.3$, $S = 2$; (b) Reference isotropic model with same S , J_{1x} and J_2 .

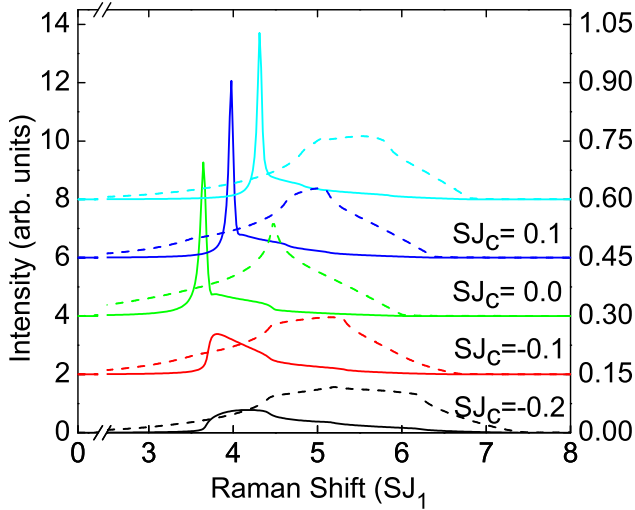


Figure 6. (Color online). J_c dependence of two-magnon Raman spectra for frustrated $J_1 - J_2 - J_c$ model $SJ_1 = 1$, $SJ_2 = 0.3$, $S = 2$. Solid lines represent B_{1g} channel and dashed lines represent A'_{1g} channel.

dependence on k_z , thus can be still treated as a 2D process. The k_z dependent magnon dispersion can be considered as an effective damping to the 2D system. Such an effective damping only affects the interacting part of the scattering cross section, and the bare part is not influenced. The magnitude of this effective damping at the $(\pi, 0)$ point is evaluated to be $\sim 8SJ_c$ for the FM J_c and $\sim SJ_c^2/(J_1 - 2J_2)$ for the AFM J_c . We then see that the damping effect is much weaker for $J_c > 0$ compared to the $J_c < 0$ systems. This explains why the resonance peak is robust for $J_c > 0$, but are suppressed when $J_c < 0$.

IV. RESULTS FOR THE $(\pi, 0)$ COLLINEAR ORDER

We also apply the same procedure to the case when the ground state has a $(\pi, 0)$ collinear AFM order. Dif-

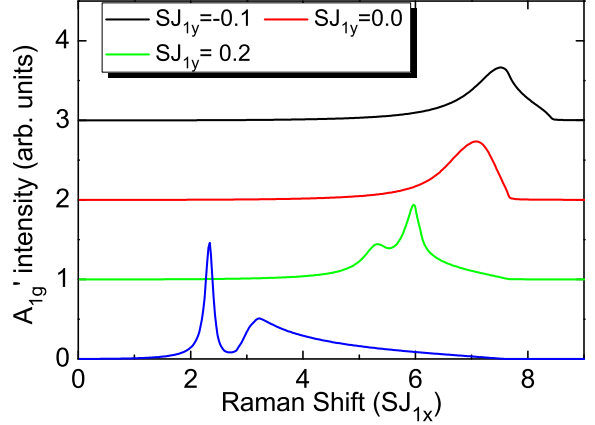


Figure 7. (Color online). J_{1y} dependence of two-magnon Raman spectra for anisotropic $J_{1x} - J_{1y} - J_2$ model with $SJ_{1x} = 1$, $SJ_2 = 0.4$, $S = 1$.

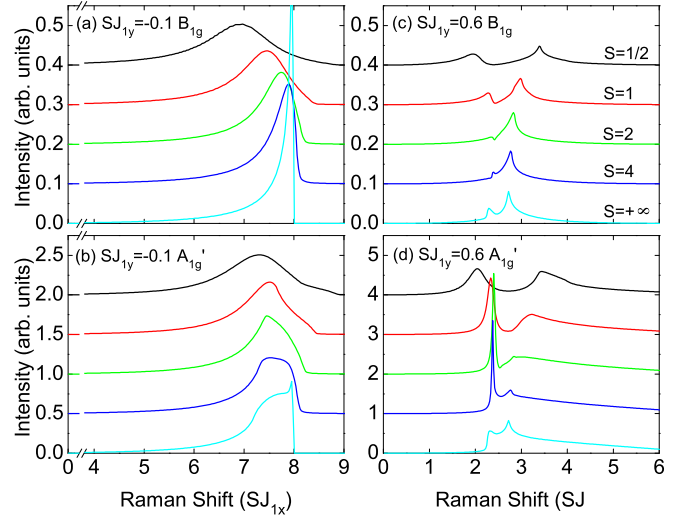


Figure 8. (Color online). S dependence of two-magnon Raman spectra for anisotropic $J_{1x} - J_{1y} - J_2$ model. (a),(b) unfrustrated model $SJ_{1x} = 1$, $SJ_{1y} = -0.1$, $SJ_2 = 0.4$; (c),(d) frustrated model $SJ_{1x} = 1$, $SJ_{1y} = 0.6$, $SJ_2 = 0.4$.

ferent from the (π, π) order, the collinear order intrinsically breaks the D_{4h} symmetry. From the discussion in the previous section, the B_{1g} and A'_{1g} channels are not separated by symmetry and share some common features in the spectra. Moreover, our calculation shows that the intensity of the A'_{1g} channel is about one order of magnitude higher than the one in the B_{1g} channel. So we will mainly focus on the spectrum of the A'_{1g} channel in this section.

The effects of frustration is shown in Fig. 7. Similar to the Néel ordered case, the frustration also pushes spectra to lower energies. But the cut-off frequency is not strongly affected by the frustration.

In a non-frustrated model $J_{1y} < 0$, the m-m interaction almost cancels the peak in bare spectra completely and develops a peak at lower energy, forming single broad

peak structure in both channels. See 8 (a)(b). As the system becomes weakly frustrated, in A'_{1g} channel the bare spectra at high energy will not be completely canceled out by the m-m interaction, resulting a two-peak structure in this channel. The low energy peak, as is resulted by scattering resonance only, is expected to vanish for large S . For highly frustrated models, the two-peak structure emerges in both channels. We can see that the two peaks survive even in the $S \rightarrow \infty$ limit, and they are pushed away from each other as $1/S$ increases, as in shown in Fig. 8 (c)(d). Note that in the A'_{1g} channel, the resonance peak becomes sharp when S is about $1 \sim 4$. This feature and its origin is very similar to the B_{1g} channel in the Néel ordered case.

Both positive and negative J_c will shift the spectra to higher frequencies. Also, as is already discussed in the previous section, the interlayer coupling J_c has the effect of a damping term to the 2D system. It is expected that sharp peak in corresponding 2D system can be damped by J_c .

V. DISCUSSIONS

A. Implications for MnBi materials

The material BaMn_2Bi_2 , which can be considered as the parent compound of the AMnBi_2 systems, is an AFM insulator with a large ordered magnetic moment $\sim 3.84\mu_B$ in each Mn ion. It has a similar structure to AMnBi_2 except that the latter are metals consisting of a layer of Dirac electrons in the AB layer.

We have calculated the two-magnon Raman spectra of a $J_1 - J_2 - J_c$ model for BaMn_2Bi_2 using the exchange parameters $SJ_1 = 21.7(1.5)$, $SJ_2 = 7.85(1.4)$, $SJ_c = 1.26(0.02)$, obtained from an inelastic neutron scattering experiment²¹. We have taken the effective spin size to be $S = 2$. The result is presented in Fig. 9. We see from the figure that a sharp resonance peak at wave number about 550 cm^{-1} is present in the B_{1g} channel. This indicates that for the model parameters taken, the effect of the m-m interaction can not be neglected, although the ordered moment is large. We note that by measuring the peak positions in the B_{1g} and A_{1g} channels as well as the cut-off frequency, one may fully determine the exchange couplings of the system. As for the Dirac materials AMnBi_2 , We expect similar Raman spectra, given that the spin dynamics is dominant by the interacting local moments. The itinerant electrons may contribute additional damping to the resonance peak in the B_{1g} channel, and may also renormalize the values of the exchange couplings via the induced RKKY interactions.

B. Iron based materials

As there is $S = 1/2$ ED calculation result for CaFe_2As_2 ($J_{1y} = -0.1J_{1x}$, $J_2 = 0.4J_{1x}$ as reported by INS measurements) available¹⁹, we make a comparison with our spectra and their 36 sites ED result, as shown in Fig. 10. Our peak position is in consistent with theirs, but the details of spectral lineshape is entirely different. What we obtain is a single broad peak structure for both polarizations. Our lineshape should be more reliable than

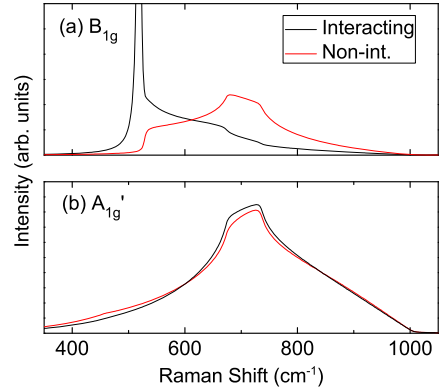


Figure 9. (Color online). Calculated two-magnon Raman spectra for BaMn_2Bi_2 with $S = 2$ and exchange parameters determined from inelastic neutron scattering experiment (see text).

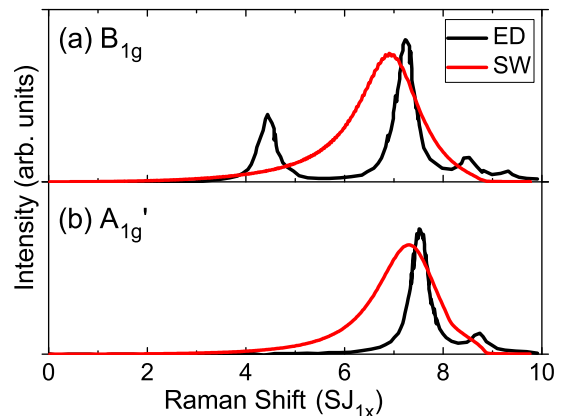


Figure 10. (Color online). Comparison of ED and spin wave approaches to two-magnon Raman spectra in anisotropic $J_{1x} - J_{1y} - J_2$ model with $SJ_{1x} = 1$, $SJ_{1y} = -0.1$, $SJ_2 = 0.4$, $S = 1/2$. (a) B_{1g} channel; (b) A'_{1g} channel.

the ED result for the presence of strong finite-size effects in the ED calculation.

Since for each Fe ion, there are two degenerate $3d$ orbitals, namely d_{xz} and d_{yz} orbitals, to be active, we can also consider an $S = 1$ spectrum for the same SJ . The spectra are shown in Fig. 8 (a)(b) (red lines). We find that one broad peak structure in both channels remains, and the peaks are slightly shifted to higher frequencies compared with the $S = 1/2$ case. The interlayer coupling J_c further shifts the peak to higher frequencies. Taking $SJ_{1x} \sim 50 \text{ meV}$ and $SJ_{1c} \sim 5 \text{ meV}$ as is reported in INS experiments, we expect a broad two-magnon peak at $7.9SJ_{1x} \sim 3185 \text{ cm}^{-1}$.

VI. CONCLUDING REMARKS

In this paper we have made a comprehensive study of two-magnon Raman spectra in $J_{1x} - J_{1y} - J_2 - J_c$ anti-ferromagnets using spin wave theory. Our treatment includes the contribution to m-m interactions at the $1/S$ order. The m-m interactions are taken into account within the ladder approximation, and the ladder diagrams are summed up exactly.

We find that for isotropic Neel ordered system, B_{1g}

Table I. FBZ

	Néel order	Collinear order
$J_c \leq 0$	$ ak_x + ak_y \leq \pi$ $ ck_z \leq \pi$	$ 2ak_x , bk_y , ck_z \leq \pi$
$J_c > 0$	$ ak_x , ak_y , ck_z \leq \pi$	$ bk_y \leq \pi$
	$ ak_x + ak_y + ck_z \leq \frac{3}{2}\pi$	$ ak_x + \frac{b}{2}k_y + ck_z \leq \frac{3}{2}\pi$

and A'_{1g} channels are well separated by symmetry. B_{1g} channel is strongly modified by m-m interaction while A'_{1g} channel is not. We predict that for large S system, in B_{1g} channel, sharp resonance peak emerges in frustrated systems when $J_c \geq 0$ and the peak is suppressed for ferromagnetic J_c . In A'_{1g} channel, a platform can be opened by J_c . For anisotropic system the B_{1g} resonance peak will tunnel into A'_{1g} channel. For collinear ordered system, we predict one peak structure for non-frustrated systems, and the peak splits due to frustration.

Our results suggest that the two-magnon Raman spectra can be used to probe the exchange anisotropy, which serves as complementary to inelastic neutron scattering. With Raman, J values can be determined by characteristic frequencies which correspond to van-Hove singularities in one-magnon DoS, since these frequencies are not shifted by m-m interactions at the $1/S$ order. Anisotropy in Néel ordered frustrated system is manifested in tunneling of the B_{1g} resonance peak into A'_{1g} channel. J 's obtained by Raman is expected to be more accurate than INS's due to its higher resolution.

Appendix A: Parameters

The definition of FBZ are shown in Table I. Note that our FBZ has spatial inversion symmetry, which is essen-

tial for simplifying the Oguchi's term $A_{\mathbf{k}}$.

The quantity $P_{\mathbf{k}}$, $Q_{\mathbf{k}}$, $A_{\mathbf{k}}$ and $B_{1234}^{(3)}$ can be written as sum of contributions from each bond: $P_{\mathbf{k}} = \sum_b J_b z_b P_{b,\mathbf{k}}$, $Q_{\mathbf{k}} = \sum_b J_b z_b Q_{b,\mathbf{k}}$, $A_{\mathbf{k}} = \sum_b J_b z_b A_{b,\mathbf{k}}$, $B_{1234}^{(3)} = \sum_b J_b z_b B_{b,1234}^{(3)}$, where b is the type of bonds, which runs over the set $\{1x, 1y, 2, c\}$, and z_b is the coordination number of bond b . The definition of $P_{b,\mathbf{k}}$, $Q_{b,\mathbf{k}}$, and $A_{b,\mathbf{k}}$ are shown in Table II. $B_{b,1234}^{(3)} = -\{\gamma_{b2-4} + \gamma_{b1-3}x_1x_2x_3x_4 + \gamma_{b1-4}x_1x_2 + \gamma_{b2-3}x_3x_4 - \frac{1}{2}[\gamma_{b2}x_4 + \gamma_{b1}x_1x_2x_4 + \gamma_{b2-3-4}x_3 + \gamma_{b1-3-4}x_1x_2x_3 + \gamma_{b4}x_2 + \gamma_{b3}x_2x_3x_4 + \gamma_{b4-2-1}x_1 + \gamma_{b3-2-1}x_1x_3x_4]\}$ for AFM bond b and $B_{b,1234}^{(3)} = \frac{1}{2}(\gamma_{b1-3} + \gamma_{b1-4} + \gamma_{b2-3} + \gamma_{b2-4} - \gamma_{b1} - \gamma_{b2} - \gamma_{b3} - \gamma_{b4})(x_2x_4 + \text{sgn}\gamma_{\mathbf{G}}x_1x_3)$ for FM bond b . Here use the notation $\gamma_{1x\mathbf{k}} = \cos k_x a$, $\gamma_{1y\mathbf{k}} = \cos k_y a$, $\gamma_{2\mathbf{k}} = \cos k_x a \cos k_y a$, $\gamma_{c\mathbf{k}} = \cos k_z a$.

It should be noticed that here we define a bond ferromagnetic, when the bond is connecting sites in the same sublattice, or antiferromagnetic otherwise. (It does not directly depend on the sign of exchange parameter of the bond.)

Values of $M_{\mathbf{k}}$ are shown in table III.

The channels $v_n(\mathbf{k})$ in Néel and Collinear ordered phase are defined in Table IV.

Matrix elements of $\hat{\Gamma}$ in Néel order is given by

$$\hat{\Gamma} = \frac{2}{N} \begin{pmatrix} \hat{X} & \hat{U} \\ \hat{U}^T & \hat{W} & \hat{V} \\ & \hat{V}^T & \hat{Z} \end{pmatrix}$$

where definition of submatrices are shown in Table V.

Table II. Definition of $P_{b,\mathbf{k}}$, $Q_{b,\mathbf{k}}$ and $A_{b,\mathbf{k}}$

	AFM bond	FM bond
$P_{b,\mathbf{k}}$	S	$-S(1 - \gamma_{b\mathbf{k}})$
$Q_{b,\mathbf{k}}$	$S\gamma_{b\mathbf{k}}$	0
$A_{b,\mathbf{k}}^a$	$\frac{1}{2\epsilon_k}(1 - \gamma_{\mathbf{k}}\gamma_{b\mathbf{k}})A_b$	$\frac{1}{2\epsilon_k}(1 - \gamma_{b\mathbf{k}})A_b$
A_b	$\frac{2}{N} \sum_{\mathbf{p}} \frac{\gamma_{\mathbf{p}}\gamma_{b\mathbf{p}} + \epsilon_{\mathbf{p}} - 1}{\epsilon_{\mathbf{p}}}$	$\frac{2}{N} \sum_{\mathbf{p}} \frac{1 - \epsilon_{\mathbf{p}} - \gamma_{b\mathbf{p}}}{\epsilon_{\mathbf{p}}}$

^a The original form of $A_{b,\mathbf{k}}$ can be written as

$$A_{b,\mathbf{k}} = \frac{2}{N} \sum_{\mathbf{p}} \frac{1}{2\epsilon_{\mathbf{k}}\epsilon_{\mathbf{p}}} [-1 + \epsilon_{\mathbf{p}} + \gamma_{\mathbf{p}}\gamma_{b\mathbf{p}} + \gamma_{b\mathbf{k}}(\gamma_{\mathbf{k}} - \epsilon_{\mathbf{p}}\gamma_{\mathbf{k}} - \gamma_{\mathbf{p}}\gamma_{b-\mathbf{k}})]$$

for AFM bond and $A_{b,\mathbf{k}} = \frac{2}{N} \sum_{\mathbf{p}} \frac{1 - \epsilon_{\mathbf{p}}}{2\epsilon_{\mathbf{k}}\epsilon_{\mathbf{p}}} (\gamma_{b\mathbf{k}-\mathbf{p}} - \gamma_{b\mathbf{k}} - \gamma_{b\mathbf{p}} + 1)$ for FM bond. It is simplified by using the equality $\sum_{\mathbf{p}} \gamma_{b\mathbf{k}}\gamma_{b\mathbf{p}} = \sum_{\mathbf{p}} \gamma_{b\mathbf{k}}\gamma_{b\mathbf{p}}$ which holds when FBZ has spatial inversion symmetry.

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Table III. Definition of $M_{\mathbf{k}}$

Polarization	Néel order	Collinear order
B_{1g}	$\sqrt{\frac{2}{N}} \frac{S}{2\epsilon_k} [J_{1x} z_{1x} (\gamma_{1x\mathbf{k}} - \gamma_{\mathbf{k}}) - J_{1y} z_{1y} (\gamma_{1y\mathbf{k}} - \gamma_{\mathbf{k}})]$	$\sqrt{\frac{2}{N}} \frac{S}{2\epsilon_k} [J_{1x} z_{1x} (\gamma_{1x\mathbf{k}} - \gamma_{\mathbf{k}}) - J_{1y} z_{1y} \gamma_{\mathbf{k}} (1 - \gamma_{1y\mathbf{k}})]$
A'_{1g}	$\sqrt{\frac{2}{N}} \frac{S}{2\epsilon_k} \{J_{1x} z_{1x} (\gamma_{1x\mathbf{k}} - \gamma_{\mathbf{k}}) + J_{1y} z_{1y} (\gamma_{1y\mathbf{k}} - \gamma_{\mathbf{k}}) + 4J_2 \times 2 \times \gamma_{\mathbf{k}} [1 - \cos(k_x a + k_y a)]\}$	$\sqrt{\frac{2}{N}} \frac{S}{2\epsilon_k} \{J_{1x} z_{1x} (\gamma_{1x\mathbf{k}} - \gamma_{\mathbf{k}}) + J_{1y} z_{1y} \gamma_{\mathbf{k}} (1 - \gamma_{1y\mathbf{k}}) + 4J_2 \times 2 \times [\cos(k_x a + k_y a) - \gamma_{\mathbf{k}}]\}$

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Table IV. Definition of the channels $v_{n\mathbf{k}}$

n	Néel	Collinear
1	$l_{\mathbf{k}}^2 \cos k_x$	$l_{\mathbf{k}}^2 \cos k_x$
2	$l_{\mathbf{k}}^2 \sin k_x$	$l_{\mathbf{k}}^2 \sin k_x$
3	$m_{\mathbf{k}}^2 \cos k_x$	$m_{\mathbf{k}}^2 \cos k_x$
4	$m_{\mathbf{k}}^2 \sin k_x$	$m_{\mathbf{k}}^2 \sin k_x$
5	$l_{\mathbf{k}}^2 \cos k_y$	$l_{\mathbf{k}}^2 \cos k_x \cos k_y$
6	$l_{\mathbf{k}}^2 \sin k_y$	$l_{\mathbf{k}}^2 \sin k_x \cos k_y$
7	$m_{\mathbf{k}}^2 \cos k_y$	$l_{\mathbf{k}}^2 \cos k_x \sin k_y$
8	$m_{\mathbf{k}}^2 \sin k_y$	$l_{\mathbf{k}}^2 \sin k_x \sin k_y$
9	$l_{\mathbf{k}} m_{\mathbf{k}} \cos k_x \cos k_y$	$m_{\mathbf{k}}^2 \cos k_x \cos k_y$
10	$l_{\mathbf{k}} m_{\mathbf{k}} \sin k_x \cos k_y$	$m_{\mathbf{k}}^2 \sin k_x \cos k_y$
11	$l_{\mathbf{k}} m_{\mathbf{k}} \cos k_x \sin k_y$	$m_{\mathbf{k}}^2 \cos k_x \sin k_y$
12	$l_{\mathbf{k}} m_{\mathbf{k}} \sin k_x \sin k_y$	$m_{\mathbf{k}}^2 \sin k_x \sin k_y$
13	$l_{\mathbf{k}} m_{\mathbf{k}}$	$l_{\mathbf{k}} m_{\mathbf{k}} \cos k_y$
14	$l_{\mathbf{k}} m_{\mathbf{k}} \cos k_z$	$l_{\mathbf{k}} m_{\mathbf{k}} \sin k_y$
15	$l_{\mathbf{k}} m_{\mathbf{k}} \sin k_z$	$l_{\mathbf{k}} m_{\mathbf{k}}$
16	$m_{\mathbf{k}}^2 \cos k_z$	$l_{\mathbf{k}} m_{\mathbf{k}} \cos k_z$
17	$m_{\mathbf{k}}^2 \sin k_z$	$l_{\mathbf{k}} m_{\mathbf{k}} \sin k_z$
18	$l_{\mathbf{k}}^2 \cos k_z$	$m_{\mathbf{k}}^2 \cos k_z$
19	$l_{\mathbf{k}}^2 \sin k_z$	$m_{\mathbf{k}}^2 \sin k_z$
20	-	$l_{\mathbf{k}}^2 \cos k_z$
21	-	$l_{\mathbf{k}}^2 \sin k_z$

Table V. Definition of submatrices in $\hat{\Gamma}$

	Néel	Collinear
\hat{X}	$-2 \begin{pmatrix} J_{1x} \hat{\mathbf{1}}_{4 \times 4} & & \\ & J_{1y} \hat{\mathbf{1}}_{4 \times 4} & \\ & & -4J_2 \hat{\mathbf{1}}_{4 \times 4} \end{pmatrix}$	$-2 \begin{pmatrix} J_{1x} \hat{\mathbf{1}}_{4 \times 4} & & \\ & 2J_2 \hat{\mathbf{1}}_{8 \times 8} & \\ & & -2J_{1y} \hat{\mathbf{1}}_{2 \times 2} \end{pmatrix}$
\hat{U}	$-2 \begin{pmatrix} J_{1x} & 0 & J_{1x} & 0 & J_{1y} & 0 & J_{1y} & 0 & 4J_2 & 0 & 0 & 0 \end{pmatrix}^T$	$-2 \begin{pmatrix} J_{1x} & 0 & J_{1x} & 0 & 2J_2 & 0 & 0 & 0 & 2J_2 & 0 & 0 & 0 & 2J_{1y} & 0 \end{pmatrix}^T$
W	$-4J_{1x} - 4J_{1y} + 8J_2 - 4 J_c $	$-4J_{1x} + 4J_{1y} - 8J_2 - 4 J_c $
\hat{V}		$-4J_c \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} J_c \leq 0$ $-2J_c \begin{pmatrix} 0 & 0 & 1 & 0 & 1 & 0 \end{pmatrix} J_c > 0$
\hat{Z}		$4J_c \begin{pmatrix} \hat{\mathbf{1}}_{2 \times 2} & \\ & \hat{\mathbf{0}}_{4 \times 4} \end{pmatrix} J_c \leq 0$ $-2J_c \begin{pmatrix} \hat{\mathbf{0}}_{2 \times 2} & \\ & \hat{\mathbf{1}}_{4 \times 4} \end{pmatrix} J_c > 0$